IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of : Confirmation No. 8224

Toru SUZUKI et al. : Docket No. 00653/01-F-011US/NT

Serial No. 09/854,528 : Group Art Unit 1731

Filed May 15, 2001 : Examiner Christopher Fiorilla

OIPE NOV 19 2003 PATENT & TRADEMARK OFFICE

ORIENTED SINTERED CERAMIC PRODUCT AND MANUFACTURING METHOD THEREOF :

RECEIVED
NOV 26 2003
TC 1700

DECLARATION UNDER 37 CFR 1.132

We, Toru Suzuki and Yoshio Sakka, the inventors named in the above-identified application, hereby declare as follows:

1. The co-authors named in the reference (HEISEI 12 SPRING MEETING, page 98) applied by the Examiner in the Office Action of May 19, 2003 are Toru Suzuki, Hideyuki Ohtuka, Yoshio Sakka, Keijiro Hiraga and Koichi Kitazawa, as apparent from the attached copy of page 98 of the reference and the attached English translation thereof.

2. Hideyuki Ohtuka, Keijiro Hiraga and Koichi Kitazawa served only as advisors to us in connection with the work reflected in this reference, and were working under our direct supervision.

3. Neither Hideyuki Ohtuka nor Keijiro Hiraga nor Koichi Kitazawa is a co-inventor of th subject matter described in this reference.

4. We further declare that all statements made herein of our own knowledge are true, and that all statements on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States

Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Tohru Suzuki

Toru Suzuki

November 12, 2003

Date

Yoshio Sakka

Yoshio Sakka

November 12, 2003

Date

4 — 14A

強磁場中スリップキャストによるアルミナの配向

(金属材料技術研究所) ○鈴木 達、大塚 秀幸、目 義雄、平賀 啓二郎
(東京大学) 北澤 宏一

[はじめに] セラミックスの作製プロセスの一つであるコロイドプロセスは粒子を液体中に分散し、それから固化成形する方法であり、粒子を微細に分散し高密度に成形するのに有効である。このプロセスに微粒子を用いることで、より微細な構造を持つセラミックスの作成が行われている。一方、近年の超伝導マグネットの発達により液体ヘリウムを使用せずに強磁場(10T 程度)を比較的簡単に得られるようになってきており、非磁性物質にも外界から及ぼされる無視できないエネルギーとして強磁場を作用させることができるようになってきた。一般に六方晶系の結晶構造では、c 軸方向と c 軸垂直方向(底面)とで結晶磁気異方性を示す。アルミナは六方晶系の結晶構造を持つが、非磁性物質で磁化率が非常に小さいためその磁気異方性は従来は無視できるものとして扱われてきた。しかし、強磁場を用いる事でこの結晶磁気異方性が無視できない量にまで大きくなるとしたら、強磁場を外界から印加する事によりアルミナの組織制御が可能になるのではないかと期待できる。そこで、本研究では粒子が良く分散して流動性の良いサスペンジョンを作製し、強磁場中でスリップキャストを行なうことで結晶が配向したアルミナを作製することを試みた。

[方法] 原料アルミナとして粒径 0.4μm(住友化学 AA-03)の粒状微粉末と 0.2μm(大明化学 TM-DAR)の粒状微粉末を用いた。水系サスペンジョンにそれぞれの粉末を分散させるため、超音波で 10 分攪拌を行ない、さらにスターラーで 12h 以上攪拌した。サスペンジョン中で粒子を安定に分散させる方法として、高分子電解質(東亜合成 アロン A-6114)を粒子表面に吸着させその電離による静電反発力と高分子そのものによる立体障害を利用する方法を用いた。このサスペンジョンを固化成形するためのスリップキャストを 10T の強磁場中で行ない、アルミナの組織制御を試みた。その後、400MPa での CIP 処理と 1400～1600℃での焼結を行なった。アルミナの配向を XRD により測定し、FE-SEM、光学顕微鏡での組織観察を行なった。

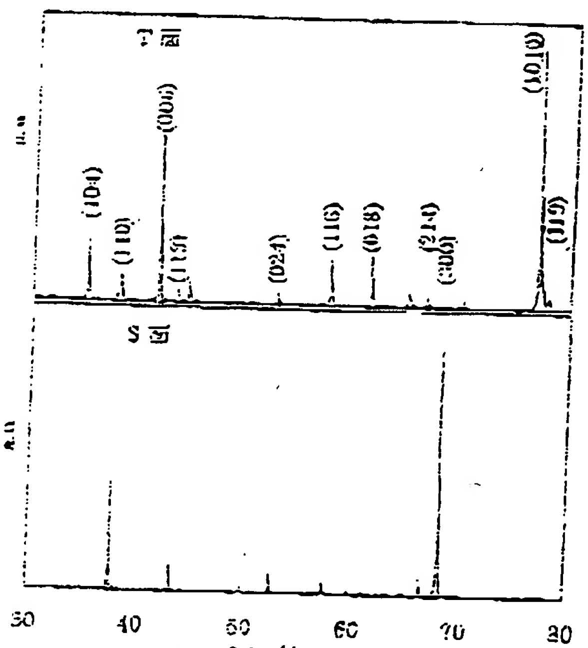
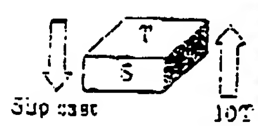


図1、焼結体磁場印加平行面と垂直面の XRD 測定結果

[結果] スリップキャストした時の底面と平行な面を T 面、垂直面を S 面とした。右図には 1600℃で焼結した試料の T 面と S 面の XRD 結果を示す。磁場印加方向は図に示したようにスリップキャスト方向と平行である。T 面では六方晶系の C 面と C 面と平行な(006)面の強度が非常に強く、また平行に近い(104)、(1010)面の強度が強く現れている。一方、S 面では c 軸と平行な(110)、(300)面の回折強度が強くなっている。これらは六方晶の c 軸が磁場印加方向と平行に配向している事を示している。図 2 に示すように 1600℃焼結体では高いアスペクト比をもつ板状組織が磁場印加方向と垂直に発達していた。これらのことから強磁場中でのスリップキャストで一軸方向に配向したアルミナの作成が可能な事が明らかになった。



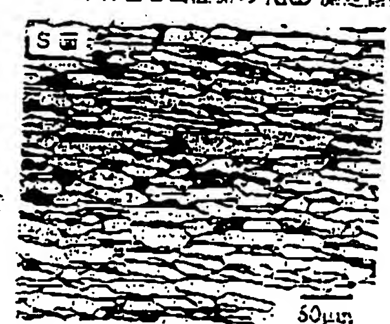


図2、磁場印加方向と平行面の組織

すずき とおる、おおつか ひでゆき、さっか よしお、ひらが けいじろう、きたざわ こういち

4－14A

Orientation of Alumina by Slip Casting in Ferromagnetic Field

(National Research Institute for Metals) Toru SUZUKI,
Hideyuki OHTUKA, Yoshio SAKKA, Keijiro HIRAGA
(Tokyo University) Koichi KITAZAWA

[Introduction]

A colloid process which is one of the processes for the manufacture of ceramics is a method where particles are dispersed in liquid, solidified and molded therefrom and it is effective for finely dispersing the particles and for molding them in a high density. Production of ceramics having finer structure has been carried out using fine particles for this process. On the other hand, as a result of the development of superconducting magnets in recent years, a ferromagnetic field (about 10 T) has been able to be achieved relatively easily without the use of liquid helium and it has been possible to apply a ferromagnetic field as an non-negligible energy which is applicable from outside even to a non-magnetic substance. Generally speaking, a hexagonal crystal structure shows a crystal magnetic anisotropy in the direction of c-axis and in the vertical direction to the c-axis (bottom). Although alumina has a hexagonal crystal structure, it is a non-magnetic substance and has a very little magnetic susceptibility whereby its magnetic anisotropy has been considered to be negligible. However, if the crystal magnetic anisotropy becomes too much

t neglect by the use of a ferromagnetic field, it is expected that a tissue control of alumina becomes possible by applying a ferromagnetic field from outside. Under such circumstances, it has been attempted to produce alumina wherein the crystals are oriented by preparing a suspension having a good fluidity where the particles are well dispersed followed by subjecting to a slip casting in a ferromagnetic field.

[Methods]

With regard to the starting alumina, granular fine powder having a particle size of 0.4 μm (AA-03; Sumitomo Chemical) and granular fine powder having a particle size of 0.2 μm (TM-DAR; Daimei Kagaku) were used. In order to disperse each of the powder materials into an aqueous suspension, stirring was carried out by means of ultrasonic wave for 10 minutes and then further carried out for not shorter than 12 hours using a stirrer. As to a method for dispersing the particles in the suspension in a stable manner, there was used a method in which a high-molecular electrolyte (Aron A-6114; Toa Gosei) was adsorbed on the surface of the particles whereby an electrostatic repulsion by its electrolysis and a steric hindrance of the high-molecular substance itself were utilized. A slip casting for the solidification and the molding of this suspension was carried out in a ferromagnetic field of 10 T whereby a tissue control of alumina was attempted. After that, a CIP treatment

at 400 MPa and a sintering at 1400-1600°C were carried out. Orientation of alumina was measured by an XRD and observation of the tissue was carried out by an FE-SEM and an optical microscope.

[Results]

A plane which was parallel to the bottom and that which was vertical thereto after the slip casting were called plane T and plane S, respectively. In the drawing on the right, results of an XRD for the planes T and S of the sample sintered at 1600°C are shown. The direction of application of a magnetic field is parallel to the slip casting direction as shown in the drawing. In the plane T, intensities of the plane C of the hexagonal system and the plane (006) parallel to the plane C were very strong while intensities of the planes (104) and (1010) nearly parallel thereto were strong. On the other hand, in the plane S, the diffraction intensities of planes (110) and (300) parallel to the c-axis were strong. Those indicate that the c-axis of hexagonal system is oriented in parallel to the direction of application of the magnetic field. As shown in Fig. 2, in a product sintered at 1600°C, a plate-like tissue having a high aspect ratio developed in a direction which was vertical to the direction of application of the magnetic field. From those, it is n w clear that production of alumina oriented in a uniaxial direction is possible by a slip casting in a ferr magnetic field.

Fig. 1 Result of an XRD Measurement of the Parallel and Vertical Planes to the Direction of Application of Magnetic Field to Sintered Substance

(upper chart) Plane T

(lower chart) Plane S

Fig. 2 Tissue of the Parallel Plane to the Direction of Application of Magnetic Field

Plane S